Selection of ferroelectric ceramics for transducers and electrical energy storage devices

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Abstract:

The selection of an optimal ferroelectric material according to the user requirements is a crucial as well as onerous task; examples of such requirements include high efficiency, sensitivity, wide operating temperature and frequency range, compact size, low cost and low loss etc. In this paper quality function deployment (QFD) in combination with multiple attribute decision making (MADM) is employed for material selection. $Pb_{(1-x)}La_x)(Zr_yTi_{(1-y)})O_3$ [PLZT (7/60/40)] (lead-based) and $(K_{0.44}Na_{0.52}Li_{0.04})$-$(Nb_{0.84}Ta_{0.1}Sb_{0.06})O_3$ (KNN-LT-LS) (lead-free) are found to be the top ranked piezoelectric ceramics for transducer applications. PLZT (7/60/40) (lead-based) and $0.7Bi_{0.5}Na_{0.5}TiO_3$-$0.2Bi_{0.5}K_{0.5}TiO_3$-$0.1(Bi_{0.5}Li_{0.5})TiO_3$ (lead-free) are found to be best materials for energy storage applications.

1. Introduction

Materials science and materials development is one among the most rapidly growing fields today with particular interest related to innovations in "functional electronic materials". Ferroelectric materials belong to most renowned families of the functional materials. These are being widely used in sensors, actuators, energy harvesting devices and many other applications. Due to their exceptionally suitable piezoelectric, pyroelectric and non-linear optical properties they have attracted the attention of researchers and technologists around the globe. A significant number of materials have been reported in this area [1-3] and these are further sub divided into two categories of lead-based and lead-free piezoelectric ceramics, primarily due to recent EU legislation restricting the use of lead [4]. The most popular systems are the lead zirconate titanate (PZT) family in the lead-based systems [1] and $(K,Na)NbO_3$ (KNN), $(Bi_{0.5}Na_{0.5})TiO_3$ (BNT) and $(Bi_{0.5}K_{0.5})TiO_3$ (BKT) among the lead-free piezoelectric ceramics. These systems are popular due to their exceptionally good piezoelectric properties as compared to other reported materials to date [5-7]. It is to be noted that PZT-based ceramics make severe negative impacts on environment [8]. KNN ceramic has some critical issues such as volatility of alkali-oxides, compositional inhomogeneity, poor densification and phase stability [9]. On the other hand, the properties of pure BNT and BKT ceramics are as good as PZT or KNN materials but their solid solutions are sufficiently good for the technological applications [9]. However, it is to be noted that all suitable or required physical properties from application point of view are rarely observed in any single material. As a result researchers are left with no other option rather than enhancing the key parameters/properties by playing with fabrication/processing variables or with compositional modifications. Here the important question is "Which composition to be improved?" and the most obvious answer is "The material which is best at present." The next point is how to judge the best composition and what parameters must be improved which can be answered by the user community. Therefore, there is a need for a user oriented approach which raises the voice of customer. One approach that is applicable for this role is quality function deployment (QFD). The concept originated in 1960s and is globally recognized after successful implementation in “Kobe shipyard of Mitsubishi Heavy Industry, Japan” [10]. Later in 1980s many companies such as General Motors, Chrysler, Digital Equipment, Hewlett-Packard, AT&T, Procter and Gamble, and Baxter Healthcare [11, 12] realized its importance and adapted this approach. This has been widely applied to various fields for numerous applications [13, 14]. It is the most famous and promising industrial engineering tools for product development and designing.

In the present study, we have used QFD for predicting weights of the material properties according to the user requirements. It has the feature to convert the verbal reasoning of customer needs to quantitative weights of material properties. Further, we are left with a few attributes (with quantitative weights) and an enormous pool of potential materials reported in literature. The selection of an optimal material from pool of alternative materials on the basis of two or more attributes/properties is a MADM problem [15]. A variety of methods are reported under MADM category. These methods include simple additive weighting (SAW), analytic hierarchy process (AHP) [16], graph theory and matrix approach (GTMA) [17], VlseKriterijumska Optimisacija I Kompromisno Resenje (VIKOR) [18], technique for order preference by similarity to ideal solution (TOPSIS) [19] and many more. These methods have some advantages and disadvantages over others. MADM models are used to select best alternative from the large number of alternatives for a set of selection criteria. Moreover these also inform the user about the degree of closeness in terms of rank index. These have been successfully applied to various fields such as manufacturing processes, social science decisions, financial decisions and engineering problems. We have found that these methods are also efficient in material selection [20-23]. We employed "Shannon entropy with TOPSIS, MDL aided-VIKOR and Pareto Optimality" methods for selection of optimal piezoelectric materials. We found that $K_{0.5}Na_{0.5}NbO_3$-$LiTaO_3$-$LiSbO_3$ (KNN-LT-LS) is one of the best piezoelectric materials in lead-free piezoelectric materials, which is also in confirmation with the experimental results. Though results predicted by these methods are reliable but it is the fact that these techniques are entirely based on data, experts opinion and do not relate the engineering goals with scientific requirements. So In present study, we propose an effective material selection approach which relates the researcher requirements with material properties. Here, we employ VIKOR with QFD weights for selection of appropriate material for two different applications namely transducers and electrical storage devices with the following objectives a) identification of parameters that a researcher should consider while synthesizing and fabricating a device; b) relations between user requirements with technical specifications or engineering characteristics, c) inter-relationships among technical specification, d) inter-relationships among user requirement or researcher goals. e) prioritization of the goals and f) selection of material for a particular application.

2. Materials and Methods

As discussed above, ferroelectric materials belong to an extensively studied family of materials. Their various compositions with different properties are widely reported in the literature. However, mere presence of the piezoelectric properties does not make all of these potential materials viable for technological applications. Many factors simultaneously govern the suitability of a piezoelectric material for different applications. These factors can be sub-divided into two categories namely ‘primary’ and ‘secondary’. Primary factors include physical properties of the material while secondary factors deals with cost, durability, toxicity, availability, ease and time of fabrication, environmental conditions etc. In this case we are much more concerned about selection of materials with optimal primary properties. Among the important material properties for piezoelectric transducers and energy storage applications the electromechanical coupling (k_p), dielectric constant (ε_r), dielectric loss ($Tan\delta$), Curie temperature (T_c) and piezoelectric coefficients (d_{33}) are reported to be critical parameters. Vital piezo-ceramics along with their properties are listed in Table 1 [24-44].

2.1. Quality Function Deployment (QFD)

QFD is evoked to step up the efficiency of product design process based on customer requirements. It is entirely based on the relationship between requirements of customer and researcher’s and engineering characteristics/material properties. In this context, a “house of quality” is prepared which connects the ‘voice’ of the customer with the technical requirements and can be considered the ‘soul’ of the QFD system.Fig.1 demonstrates a typical house of quality for the problem understudy. It is a matrix ($i \times j$) which has following building blocks (sub-matrix):

- **Whats:** This includes expectations of customers/researchers, in terms of what they are looking for in the particular product to be studied. Examples can include cost, life, working temperature, frequency range, sensitivity etc. These are to be listed along rows in the left hand side of the house of quality. The product/device has to be fabricated in order to satisfy these expectations.

- **Hows:** This consists of the prime technical parameters (such as electromechanical coupling (k_p), dielectric constant (ε_r), dielectric loss ($Tan\delta$) piezoelectric coefficients (d_{33}), Curie temperature (T_c)) which are responsible for satisfying the customer needs.
- **Hows-correlation (roof):** This enlightens the inter-dependence of technical parameters.
- **Planning matrix:** This provides information about the customer perceptions. It is used to priorities the customer needs to fabricate an eminent product or device. All of the "whats" are given priorities *(Pr)* on a scale of 1-5 (1-less important, 2-important, 3-much more important, 4-very important and 5-most important) as per the customer perception and opinion.
- **Inter-relationship matrix:** This explains the relationship between "Hows" and "Whats" in terms of a correlation index. The correlation index is an appropriate set of numbers for assigning importance. These are filled as per the dependence of the customer requirements on technical characteristics (material properties) as 1-very weak, 3- weak, 5-modrate, 7-stong, 9- strongest. It also takes account of correlation among material properties, i.e. hows.
- **Weights *(W_j)*:** This gives the overall quantitative weightage of material properties with respect to the features described by customer/researchers. It is calculated as

$$W_j = \sum_{i=1}^{n} \mathrm{Pr}_i \times ID \times \text{correlation index} \tag{1}$$

Where, *ID* is improvement driver which shows benefit (+1) and loss (-1) criteria for customer requirements.

2.2. VIKOR method

The VIKOR method is a compromise approach MADM model [18]. The analysis of VIKOR is highly accurate [45] and provide close to a real solution. It makes the use of utility weight, thus enabling different users to apply expert opinion. The normalization norms used in VIKOR are linear. The calculation of the VIKOR index involves the following steps;

Step 1: Determination of ideal and negative ideal solution;

The ideal solution f^* and the negative ideal solution f^- are determined as:

$$f^* = \{(\max f_{ij}, j \in J) or (\min f_{ij}, j \in J^{'})\} \quad (2)$$

$$f^- = \{(\min f_{ij}, j \in J) or (\max f_{ij}, j \in J^{'})\} \quad (3)$$

where f_{ij} is the j^{th} property of i^{th} material and J corresponds to benefit criteria and $J^{'}$ corresponds to cost criteria.

Step 2: Calculation of utility measure and regret measure;

$$S_i = \sum_{j=1}^{n} W_j \frac{(f_j^* - f_{ij})}{(f_j^* - f_j^-)}; \forall i \quad (4)$$

$$R_i = Max_j \left[W_j \frac{(f_j^* - f_{ij})}{(f_j^* - f_j^-)} \right]; \forall i \quad (5)$$

where S_i and R_i represent the utility measure and regret measure respectively and W_j is the relative weight assigned to the j^{th} property.

Step 3: Determination of VIKOR index;

$$Q_i = v\left[\frac{S_i - S^*}{S^- - S^*}\right] + (1-v)\left[\frac{R_i - R^*}{R^- - R^*}\right]; \forall i \tag{6}$$

where, Q_i represents the i^{th} material VIKOR value, v is the group utility weight, it is generally considered as 0.5 (unsupervised) and ;

$$S^* = Min_i(S_i); \tag{7}$$

$$S^- = Max_i(S_i); \tag{8}$$

$$R^* = Min_i(R_i); \tag{9}$$

$$R^- = Max_i(R_i); \tag{10}$$

The material with the least (lowest?) value of VIKOR index Q_i is preferred.

3. Results and Discussions

Materials science and engineering covers a broad range of multidisciplinary areas starting from physics, chemistry and leading up to decision-making, designing, economics and marketing (in short, Industrial Engineering). A lack of communication and limited understanding of requirements and interrelationships between different fields is one of the biggest hurdles in development of such materials and devices. There is therefore a need of an approach which can relate all concerned disciplines and answer all questions pointed above. In this context, QFD plays a vital role and successfully implemented in the present study.

The present study focuses on selection of piezoelectric materials with optimal properties for transducer and electrical energy storage applications. All properties such as k_p, ε_r, T_c, $Tan\delta$ and d_{33} have their own importance for various piezoelectric applications and have different priorities. Piezoelectric constant shows an ability of material to produce a high electrical field on application of mechanical strain or vice-versa, which is often a key parameter in deciding material for actuator and sensor applications. On the other hand, for energy storage applications, it is nothing more than the piezoelectric noise (unnecessary vibrations), which reduces the efficiency of the system. Similarly the dielectric constant is the essence of the ability of a material to store electrical energy and $Tan\delta$ shows inherent dissipation of stored electrical energy. These are highly significant figures of merit in case of energy storage as compared to transducer applications. k_p is the conversion efficiency of the material; which is an important feature of transducer materials but unfortunately of almost negligible importance in case of energy storage devices. Last but not the least T_c defines the temperature domain for which a device can be safely and efficiently operated.

In order to assign relative weights to above mentioned properties, we have shortlisted the user requirements (whats) and house of quality are formed for both the applications under study. Customers and researchers highlighted the requirement of a compact transducer device with high fatigue life, sensitivity, efficiency, working temperature and frequency range at low cost. Parallel investigations for energy storage devices highlighted the need for a compact design with high energy density working temperature and frequency range and efficiency at low cost and piezoelectric noise. Based on user/researcher's needs all "whats" are prioritized and an interrelationship matrix is obtained. Once the matrix is formed the weights are calculated using eq.1. Tables 3 and 4 summarize the calculation for weights (house of quality) for all the properties under study. Fig. 2 illustrates the variation of weights (percentage) of all properties for both applications understudy. It clearly shows that there can be huge variation in weightage of material properties for two different devices. Priority order for transducer is $d_{33}>T_c> k_p>Tan\delta>\varepsilon_r$ and for energy storage application $\varepsilon_r>Tan\delta>d_{33}>T_c> k_p$ respectively.

The weights (calculated using QFD) are multiplied with material properties and rank indices are obtained using VIKOR for the materials understudy. The rank index and corresponding ranks calculated for piezoelectric transducer and energy storage applications are shown in Table 1 and Table 2 respectively. PLZT (7/60/40) is found to be at top for both the application. Though it has limitation of working temperature range due to low Curie temperature as compared to the highest ranked lead-free members in the lists, it is able to attain the highest ranking position because of the exceptionally values of all other properties for these devices. It is to be noted (Table 1) that KNN-LT-LS is top lead-free material for piezoelectric transducer applications. It has been experimentally investigated and rated very high among top candidates for this application [46]. Based on our results also, it is advisable to explore KNN-based families more for piezoelectric transducer applications as most of the members of this family are among the top ranked among the lead-free piezoelectric. For energy storage applications the situation is a little different. 0.7BNT-0.2BKT-0.1($Bi_{0.5}Li_{0.5}$)TiO_3 and 0.92BNT-0.08BT+0.3 wt % MnO (Table 4) has secured top positions among the lead-free families. NBT-KBT-LBT and $BaTiO_3$ has achieved the third and fourth rank in the same group. These four are studied intensively for the storage applications in both bulk as well as thin films form [37, 39, 40, 43]. $K_{0.5}Na_{0.5}NbO_3$ is found at the bottom in the material pool under study. We suggest the aforesaid families should be explored or modified in order to have promising material properties and highly efficient devices as per the standards of user. The present study is one of the first attempts to focus on unforeseen importance of industrial engineering in material science.

4. Conclusions

QFD incorporation with VIKOR is employed for selection of ferroelectric ceramics for transducer and energy storage applications. PLZT (7/60/40) is found to be best material for both applications. Among the lead-free ferroelectrics, KNN-LT-LS and 0.7BNT-0.2BKT-0.1$(Bi_{0.5}Li_{0.5})TiO_3$ are found to be best alternatives for transducer and energy storage applications. $K_{0.5}Na_{0.5}NbO_3$ is found at the bottom in the material pool under study. Physical properties for these material are weighted as $d_{33}>T_c>k_p>Tan\delta>\varepsilon_r$ for transducer and $\varepsilon_r>Tan\delta>d_{33}>T_c>k_p$ for energy storage applications respectively.

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Figures Captions:

Figure 1: The house of quality.

Figure 2: Properties weights for transducers and electrical energy storage applications

Table 1: Ferroelectric materials, physical properties and their corresponding rank for transducer applications

Rank Index	**Rank**	**Material**	ε_r	**Tanδ**	k_p	d_{33} **(pC/N)**	T_c **(°C)**
0	1	PLZT(7/60/40)[23]	2590	0.019	0.72	710	140
0.097515	2	PLZT(8/65/35) [23]	3400	0.03	0.65	682	105
0.255479	3	KNN-LT-LS [43]	1650	0.024	0.48	340	266
0.323984	4	KNN-$LiSbO_3$ (5%)[35]	1288	0.019	0.5	283	392
0.437712	5	KNN-Li (7%)[33]	950	0.084	0.45	240	460
0.460671	6	KNN-$LiNbO_3$ (6%)[29]	500	0.04	0.42	235	460
0.526581	7	0.7BNT-0.2BKT-0.1$(Bi_{0.5}Li_{0.5})TiO_3$ [39, 34]	1900	0.044	0.368	231	290
0.528129	8	NBT-KBT-LBT [38]	1550	0.034	0.401	216	350
0.574197	9	KNN-$LiTaO_3$ (5%) [30]	570	0.04	0.36	200	430
0.61381	10	KNN-Li3%; Ta20% [40]	920	0.024	0.46	190	310
0.679242	11	NBT-KBT-BT [24]	770	0.034	0.367	183	290
0.71042	12	NBT-KBT-BT (MPB) [24]	730	0.02	0.33	173	290
0.717186	13	$BaTiO_3$ [34]	1700	0.01	0.36	190	115
0.724562	14	0.92BNT-0.08BT+0.3 wt % MnO [42]	1596	0.008	0.364	153	260
0.77785	15	$(K_{0.5}Na_{0.5})NbO_3$ (HP) [25, 26]	500	0.2	0.46	127	420
0.820247	16	BBT-KBT90 [1]	837	0.05	0.23	140	297
0.820363	17	NBT-KBT-BT [24]	820	0.03	0.162	145	302
0.840154	18	$BaTiO_3$-$CaTiO_3$-Co [37]	1420	0.005	0.31	150	105

0.907845	19	SBT-KBT85 [1]	1000	0.05	0.16	120	250
0.910125	20	SBT-KBT90 [1]	870	0.04	0.15	110	296
0.957714	21	BBT-KBT80 [1]	630	0.04	0.15	95	290
1	22	$(K_{0.5}Na_{0.5})NbO_3$ [32]	290	0.4	0.35	80	420

Table 2: Ferroelectric materials, physical properties and their corresponding rank for energy storage applications.

Rank Index	Rank	Material	ε_r	Tanδ	k_p	d_{33} (pC/N)	T_c (°C)
0.000	1	PLZT(7/60/40) [23]	2590	0.019	0.72	710	140
0.006	2	0.7BNT-0.2BKT-0.1($Bi_{0.5}Li_{0.5}$)TiO_3 [39, 34]	1900	0.044	0.368	231	290
0.097	3	0.92BNT-0.08BT+0.3 wt % MnO [42]	1596	0.008	0.364	153	260
0.136	4	NBT-KBT-LBT [38]	1550	0.034	0.401	216	350
0.163	5	$BaTiO_3$ [34]	1700	0.01	0.36	190	115
0.232	6	KNN-LT-LS [43]	1650	0.024	0.48	340	266
0.263	7	$BaTiO_3$-$CaTiO_3$-Co [37]	1420	0.005	0.31	150	105
0.270	8	PLZT(8/65/35) [23]	3400	0.03	0.65	682	105
0.300	9	KNN-$LiSbO_3$ (5%) [35]	1288	0.019	0.5	283	392
0.346	10	SBT-KBT85 [1]	1000	0.05	0.16	120	250
0.360	11	SBT-KBT90 [1]	870	0.04	0.15	110	296
0.395	12	NBT-KBT-BT [24]	820	0.03	0.162	145	302
0.435	13	BBT-KBT90 [1]	837	0.05	0.23	140	297
0.447	14	KNN-Li (7%) [33]	950	0.084	0.45	240	460
0.478	15	KNN-Li3%; Ta20% [40]	920	0.024	0.46	190	310
0.477	16	BBT-KBT80 [1]	630	0.04	0.15	95	290
0.521	17	NBT-KBT-BT (MPB) [24]	730	0.02	0.33	173	290
0.536	18	NBT-KBT-BT [24]	770	0.034	0.367	183	290

0.555	19	KNN-$LiTaO_3$ (5%) [30]	570	0.04	0.36	200	430
0.613	20	KNN-$LiNbO_3$ (6%) [29]	500	0.04	0.42	235	460
0.760	21	$(K_{0.5}Na_{0.5})NbO_3$ (HP) [25,26]	500	0.2	0.46	127	420
1.000	22	$(K_{0.5}Na_{0.5})NbO_3$ [32]	290	0.4	0.35	80	420

Table 3: House of quality matrix for transducer applications, d_{33} in pC/N, T_c in °C.

		Improvement Driver	Priority	ε_r	Tanδ	k_p	d_{33}	T_c
				Technical Requirements (Attributes)				
Customer Requirements	**1. Fatigue life**	1	4	1	1	1	9	7
	2. Sensitivity	1	4	1	3	9	9	5
	3. Working temperature	1	5	7	7	5	7	9
	4. Efficiency	1	4	1	1	9	7	1
	5. Working frequency	1	3	7	9	3	5	7
	6. Size	-1	5	1	3	5	1	5
	7. Cost	-1	1	7	3	5	1	3
			Weight	56	64	80	144	90

Table 4: House of quality matrix for energy storage applications, d_{33} in pC/N, T_c in °C.

					Technical Requirements (Attributes)			
		Improvement Driver	**Priority**	**ε_r**	**Tanδ**	**k_p**	**d_{33}**	**T_c**
Customer Requirements	**1. Energy density**	1	5	9	7	1	3	5
	2. Working frequency	1	3	5	9	1	5	7
	3. Piezoelectric noise	-1	4	1	7	1	1	3
	4. Efficiency	1	4	9	9	7	7	5
	5. Working temperature	1	3	7	7	5	5	9
	6. Size	-1	4	1	3	1	1	5
	7. Cost	-1	2	1	3	1	1	9
			Weight	107	73	41	63	43